## Superconducting atomic contacts under microwave irradiation

M. Chauvin, P. vom Stein, H. Pothier, P. Joyez, M. E. Huber, D. Esteve, and C. Urbina Quantronics Group, Service de Physique de l'État Condensé (CNRS URA 2464), DSM/DRECAM, CEA-Saclay, 91191 Gif-sur-Yvette Cedex, France
Department of Physics, University of Colorado at Denver, Denver, Colorado 80204, USA (Dated: February 6, 2008)

We have measured the effect of microwave irradiation on the dc current-voltage characteristics of superconducting atomic contacts. The interaction of the external field with the ac supercurrents leads to replicas of the supercurrent peak, the well known Shapiro resonances. The observation of supplementary fractional resonances for contacts containing highly transmitting conduction channels reveals their non-sinusoidal current-phase relation. The resonances sit on a background current which is itself deeply modified, as a result of photon assisted multiple Andreev reflections. The results provide firm support for the full quantum theory of transport between two superconductors based on the concept of Andreev bound states.

PACS numbers: 74.50.+r, 74.25.Fy, 74.45.+c, 74.78.Na, 73.63.-b

A thorough and unifying view of superconducting electrical transport emerged in the last fifteen years in the framework of mesoscopic superconductivity. It is based on the concept of Andreev reflection, the microscopic process which couples the dynamics of electrons and holes. In particular, the Josephson currents flowing between two weakly coupled superconductors are described as arising from Andreev bound states forming in each conduction channel of the coupling structure [1]. The theory predicts the time-dependent current through a voltage biased short single conduction channel of arbitrary transmission probability [2], and in particular the interplay between these ac Josephson currents and a microwave external signal [3]. Although ac supercurrents have been known and detected since the early days of Josephson circuits [4], this modern view has the advantage of being completely general as it applies to all possible coupling structures, which can always be decomposed, at least in principle, into a set of independent channels. In this Letter we present a test of these predictions carried out on single atom contacts between two superconducting electrodes [5]. These contacts are model systems which allow for a direct comparison of theory and experiment, as one can vary and measure [6] their "mesoscopic PIN", i.e. the set of transmission coefficients  $\{\tau_i\}$  characterizing their conduction channels.

In a single short channel of transmission  $\tau$  between two reservoirs with superconducting phase difference  $\delta$ , two Andreev states contribute to the Josephson coupling. They have energies  $E_{\pm}\left(\delta,\tau\right)=\pm\Delta\sqrt{1-\tau\sin^2(\delta/2)}$  lying inside the superconducting gap extending from - $\Delta$  to  $\Delta$ . At a given  $\delta$  the two states carry opposite currents  $I_{\pm}\left(\delta,\tau\right)=(1/\varphi_0)\,\partial E_{\pm}/\partial\delta$ , where  $\varphi_0=\hbar/2e$  is the reduced flux quantum, and the net current through the channel results from an imbalance of their occupation numbers. For a perfect voltage bias V the phase evolves in time according to  $\delta\left(t\right)=\omega_J t$  where  $\omega_J=V/\varphi_0$  is the Josephson frequency. Because the current-phase re-

lation of each state is periodic, there are ac supercurrents at the Josephson frequency and all its harmonics, and the current can be written as a Fourier series  $I(V,\tau,t)=\sum_{m}I_{m}(V,\tau)e^{im\omega_{J}t}$  [7]. The sine components arise from the adiabatic evolution of the system on the ground Andreev state, whereas the cosine components originate in non-adiabatic (Landau-Zener) transitions between the levels induced by the dynamics of the phase. These cosine terms become sizeable only for highly transmitting channels, and lead in particular to a dc current at finite voltage. This perfect voltage-bias, non-adiabatic theory, explains quantitatively [6], in terms of multiple Andreev reflections (MAR) [8], the strong current non-linearities known as the "subgap structure" observed at finite voltage in all kinds of SNS structures.

In the experiments presented here, we monitor the modifications under microwave irradiation of the dc current-voltage characteristics of voltage biased aluminum atomic contacts obtained using microfabricated break junctions [9]. The principle of the experimental setup is shown schematically in Fig. 1. The break junctions are embedded into a biasing circuit of low impedance (the so-called "environmental impedance") designed to approach the perfect voltage bias condition assumed in the theory. The contact is characterized by its critical current  $I_0(\{\tau_i\})$ , typically a few tens of nA. It is placed in series with a microfabricated resistor r and this combination is shunted by a microfabricated capacitor C and a surface mounted resistor R. In practice we have used two setups, which differ essentially in the way the current through the atomic contact is measured. In the first type (A), the current is measured by means of an array of 100 dc SQUIDs [10], as described in [11]. In the second type (B), the current is obtained through the voltage drop across the resistor r, directly measured with low-noise voltage amplifiers [12]. Details can be found in [13]. Both setups give essentially the same results.

A typical I(V) of a one atom aluminum contact, with

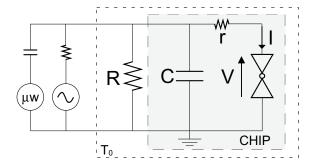


FIG. 1: Simplified schematics of the electromagnetic environment seen by the atomic contact (double-triangle symbol). Dashed grayed box shows the microfabricated on-chip environment. Dotted box shows components cooled down to base refrigerator temperature  $T_0$ . A low-frequency voltage source coupled through a large resistor provides a low-frequency current bias. Microwaves are injected through a small coupling capacitor.

no applied microwaves, is shown in Fig. 2. The strong non-linearities arising at the thresholds  $V = 2\Delta/ne$  of the different MAR processes allow to determine the gap  $\Delta$  [14] and the full  $\{\tau_i\}$  [6]. At small scale (inset of Fig. 2), the dc Josephson current manifests itself as a peak with a finite width. This physics is well-understood as due to phase fluctuations caused by the noise in the environmental impedance supposed to be at a finite temperature  $T_e$ . The theory [15], developed initially for a purely resistive environment, is based on the solution of a Langevin equation for the dynamics of the phase, which diffuses along the Josephson potential. It has been thoroughly checked experimentally in the case of tunnel junctions [11], and for structures containing only channels of small or intermediate transmission [16]. Note that this is an adiabatic theory, as it does not include the effect of Landau-Zener transitions to the excited Andreev levels that are essential to explain the experimental results in the case of highly transmitting channels [16]. The theory has been extended to the case of structures containing ballistic channels [17] and to deal with more general environments [18]. The important fact is that the size of the supercurrent peak diminishes quite rapidly with the ratio between the Josephson energy  $\varphi_0 I_0$  and the thermal energy  $k_BT_e$  available in the dissipative elements of the environment. As shown in the inset of Fig. 2, the agreement between the experimental data and the calculated phase diffusion curves using the independently measured values of  $\{\tau_i\}$ , r, C and R is excellent. However, in the present experiments the environment temperature extracted through this analysis was always significantly above that of the refrigerator [19].

When microwaves are applied the whole I(V) is deeply modified. As shown in Fig. 3, sharp resonances appear at well defined voltages which scale with the microwave fre-

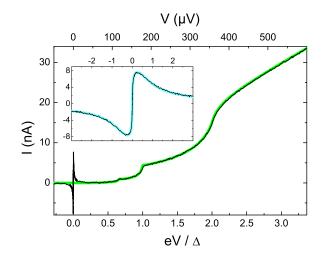


FIG. 2: (color online). Full lines: current-voltage characteristic of an Al atomic contact measured in type B setup at refrigerator temperature  $T_0=20$  mK. Grayed line: best fit using zero temperature MAR theory, obtained for PIN  $\{0.389, 0.238, 0.055\}$  and  $\Delta=178.2~\mu\text{eV}$ . Inset: zoom on the supercurrent peak. Grayed line: best fit using environment temperature  $T_e=133$  mK in phase diffusion theory.

quency  $\omega$ . The amplitude of these resonances, and of the supercurrent peak itself, oscillates with the amplitude of the microwave field. The non-adiabatic theory [2] has been extended [3] to consider a perfect voltage bias containing both a constant component V and an oscillating one  $A\cos(\omega t)$ , in which case the phase evolves in time according to  $\delta(t) = \omega_J t + 2\alpha \sin(\omega t)$  where  $\alpha = A/(2\varphi_0\omega)$ . The time-dependent current becomes [3]

$$I(V, \alpha, \tau, \omega, t) = \sum_{m,n} I_m^n(V, \tau, \alpha, \omega) e^{i[m\omega_J + n\omega]t}.$$
 (1)

In this case of perfect voltage bias, the dc component can be explicitly decomposed into a continuous background  $I_0^0(V, \tau, \alpha, \omega)$  (which for  $\alpha = 0$  corresponds to the MAR current), plus a sum of singularities  $I_m^n(V,\tau,\alpha,\omega)\delta\left(V-\frac{n}{m}\varphi_0\omega\right)$  which correspond to the well known Shapiro resonances [20] arising from the beatings between the Josephson ac currents and the external microwave probe when their frequencies are commensurate  $(m\omega_{J}=n\omega)$ . For a contact containing only low and intermediate transmitting channels (all  $\tau$ 's< 0.5), the predicted current-phase relation is almost sinusoidal and the m=1 component is the only sizeable one in the supercurrent. Therefore, like in the well-known case of tunnel junctions, Shapiro resonances appear centered at integer multiples of the Josephson voltage  $V_J = \varphi_0 \omega$  determined by the external frequency, as shown in the lower panel of Fig. 3. Obviously, the shape of the resonances cannot be understood within the constant bias theory [3] which does not allow for phase fluctuations. However, as shown by the underlying graved line in Fig. 3, the shape and size

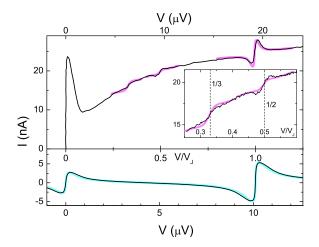


FIG. 3: (color online). Full lines: I(V)s measured under microwave excitation for two different contacts, refrigerator temperature  $T_0=20$  mK. Middle axis: voltage in Josephson voltage units ( $V_J=\varphi_0\omega$ ). Upper and bottom axis: voltage in  $\mu$ V. Upper panel: contact on a type A sample, PIN  $\{0.992,0.279,0.278\}$ ,  $\Delta=177~\mu{\rm eV}$ ,  $\alpha=0.43$ ,  $\omega/2\pi=9.3156$  GHz. Inset: zoom on the small Shapiro resonances at  $V/V_J=1/3,1/2$ . Grayed lines, predictions of the mapping model with temperature  $T_e=200$  mK. Lower curve: Same sample as in Fig. 2, but different run and contact with PIN  $\{0.573,0.233,0.037\}$ ,  $\Delta=179.7~\mu{\rm eV}$ ,  $\alpha=0.86$ ,  $\omega/2\pi=4.892$  GHz. Grayed line: phase diffusion theory [18] with environment temperature  $T_e=120$  mK.

of these resonances can be perfectly accounted for using the theory by Duprat and Levy Yevati [18] who have extended the Fokker-Planck treatment of [15] to include the microwave drive. For these small transmissions there is essentially no MAR current in the voltage range of the Shapiro resonances, and this adiabatic theory works well. For small transmissions the amplitude of the n-resonance varies with the reduced microwave probe amplitude  $\alpha$  basically as a Bessel function of order n (data not shown), which allows for a calibration of the microwave driving current. In the top panel of Fig. 3 we show the results on a contact containing a highly transmitting channel. The most important qualitative fact is the appearance of small resonances at fractional multiples of the Josephson voltage. These so-called fractional Shapiro resonances are a direct consequence of the deviation of the currentphase relationship from a pure sine function. The resonances occur at voltages for which there is an important MAR current, which is itself modified by the microwave field, and the current cannot be decomposed into two distinct contributions as before. As there exists no theory dealing with this situation of non-adiabatic phase diffusion in presence of microwaves, we have developed an empirical model in which the resonances are viewed as replicas of the supercurrent peak. We take into account the effects of the environment by mapping the dynamics of the phase around each resonance into the dynamics

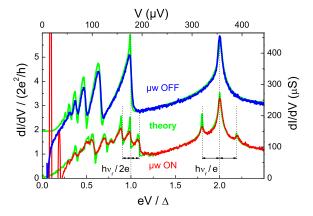


FIG. 4: (color online). Full lines: measured differential conductance of contact with PIN  $\{0.696, 0.270, 0.076\}$  and  $\Delta=177.6~\mu\text{eV}$ . Upper curve (shifted upwards by 150  $\mu\text{S}$ ): no microwaves. Lower curve: under microwave irradiation with  $\alpha=0.70,~\omega/2\pi=8.2935$  GHz. Grayed lines: predictions of PAMAR theory, with no adjustable parameters. The theory includes neither the negative contribution of the Josephson peak at low voltages, nor the Shapiro resonances.

around zero voltage in absence of microwaves. In other words, we suppose that the phase fluctuates around the deterministic dynamics imposed by an hypothetical perfect voltage bias (both dc and microwave). Under this hypothesis, the system is governed by a Langevin equation similar to the one describing the dynamics in absence of microwaves, differing simply by an offset in voltage  $\frac{n}{m}V_J$  and the following scaling of the parameters: For each  $\frac{n}{m}$  resonance, the Josephson critical current is replaced by its maximum amplitude predicted by the perfect bias, non-adiabatic theory [3] for the measured  $\{\tau_i\}$ , and most importantly, the environment temperature  $T_e$ has to be replaced by an effective temperature  $mT_e$  [13]. This means that fractional Shapiro resonances are very rapidly washed out by thermal fluctuations [21], as compared to the integer resonances. The underlying graved lines of the upper panel in Fig. 3 are the predictions of this mapping approach, the environment temperature being the only adjustable parameter. The best fit to the data is obtained assuming an environment temperature of  $T_e = 200$  mK instead of the actual temperature read by the thermometers  $T_0 = 20$  mK. A linear background term has also been added to account, at least partially, for the background current on which the Shapiro resonances superimpose. The model describes the general trends of the experimental results. In particular the amplitude of the integer resonances as a function of the amplitude of the microwave field are quite well accounted for (data not shown). However, the amplitude of the fractional resonances is too small to make a quantitative comparison with theory.

For the typical microwave frequencies (< 12 GHz) and amplitudes used here, the Shapiro resonances are ob-

served over a small voltage range ( $|eV| < 0.2\Delta$ ). However, at larger voltages there is still a large effect of the irradiation on the I(V). Figure 4 shows a comparison of the measured and calculated differential conductance dI/dV, in presence of microwaves. With no microwaves, the onsets of the different MAR processes of Fig. 2 appear as peaks on the differential conductance curve. In presence of microwaves, satellite peaks appear around them, at voltages  $V = (2\Delta \pm m\hbar\omega_r)/2ne$ . They correspond to the absorption or emission of m photons during the MAR process which transfers n electronic charges, i.e. to photon-assisted MAR processes (PAMAR). The experimental results are very well reproduced by the dc component  $I_0^0(V, \tau, \alpha, \omega)$  of Eq. 1, with no adjustable parameters. Note that the theory does not consider the effect of thermal fluctuations of the phase. For all the contacts we have measured, the agreement between theory and experiment is as good as shown in Fig. 4. Although these multiphoton processes have been already observed [22] and identified [23], to our knowledge this is the first direct quantitative comparison between theory and experiment.

In conclusion, the ability of tuning and measuring the transmission of the few channels accommodated by atomic contacts allows to compare, with no adjustable parameters, experimental results with the predictions of the modern theory of the Josephson effect. The observation of fractional Shapiro resonances is clear indication of the occurrence of supercurrents at harmonics of the Josephson frequency in contacts of large transmission. Furthermore, we find quantitative agreement between our results and the predictions of the theory of photon-assisted multiple Andreev reflections. The results illustrate the power of this modern view, which is able to describe both dissipative and non-dissipative currents, simply in terms of occupation of Andreev levels.

This work was supported by the EU Network DIENOW. We acknowledge technical assistance of P.F. Orfila and P. Senat, and important discussions with J. C. Cuevas, R. Duprat, G. Rubio Bollinger, A. Martín Rodero and A. Levy Yeyati, who also provided us with their computer codes.

- A. Furusaki and M. Tsukada, Solid State Commun. 78, 299 (1991).
- [2] D. Averin and A. Bardas, Phys. Rev. Lett. **75**, 1831 (1995); J. C. Cuevas, A. Martín-Rodero, and A. L. Yeyati, Phys. Rev. B **54**, 7366 (1996); E. N. Bratus' *et al.*, Phys. Rev. B **55**, 12666-12677 (1997).
- [3] J. C. Cuevas et al., Phys. Rev. Lett. 88, 157001 (2002).
- [4] I. K. Yanson, V. M. Svistunov, and I. M. Dmitrenko, Zh. Eksp. Teor. Fiz. 48, 976 (1965) translated in Sov. Phys. JETP 21, 650 (1965); I. Giaever, Phys. Rev. Lett. 14, 904 (1965); D. N. Langenberg et al., Phys. Rev. Lett. 15,

- 842 (1965).
- [5] N. Agraït, A. Levy Yeyati, and J. M. van Ruitenbeek, Phys. Rep. 377, 81 (2003).
- [6] E. Scheer et al., PRL 78, 3535 (1997).
- [7] Under voltage bias the Andreev levels acquire a finite width due to the coupling with the continuum of states beyond the gap, and the theory only provides  $I_m(V, \tau)$  as integrals that have to be evaluated numerically [2].
- [8] M. Octavio et al., Phys. Rev. B 27, 6739 (1983).
- [9] J. M. van Ruitenbeek *et al.*, Rev. Sci. Inst. **67**, 108 (1996).
- [10] R. P. Welty and J. M. Martinis. IEEE Trans. Mag. 27, 2924 (1991); M. E. Huber et al., Appl. Supercond. 5, 425 (1998).
- [11] A. Steinbach et al., Phys. Rev. Lett. 87, 137003 (2001).
- [12] Both types of samples are fabricated on metallic substrates(ground plane). The break junctions are tailored in a 100 nm thick Al film. In samples of type A, the onchip resistor  $r \sim 7.5 \Omega$  is a 165  $\mu$ m-long, 28  $\mu$ m-wide, 60 nm-thick Au:Cu (3:1 in weight) film. The capacitor  $C \sim 100 \text{ pF}$  is formed between a 4 mm<sup>2</sup> square of the same film and the substrate. The dielectric is a 1.6  $\mu$ mthick polyimide layer. Shunt was  $R \sim 30 \Omega$ . In samples of type B, designed for four-point measurements, there are four identical copies of each microfabricated element. The 70  $\mu$ m-long, 0.5  $\mu$ m-wide on-chip resistors  $r \sim 40 \Omega$ are tailored in a 30 nm-thick Au film. The top plates of the capacitors  $C \sim 92~\mathrm{pF}$  are 200 nm-thick Au electrodes. The shunt resistor was  $R \sim 50~\Omega$ . Values of resistances and capacitances where measured independently and varied within 1% from sample to sample.
- [13] P. vom Stein et al., in "Quantum Information and Decoherence in Nanosystems", ed. D. C. Glattli, M. Sanquer and J. Trăn Thanh Văn, Thě Giði Publishers, Hanoi (2004); M. Chauvin, PhD Thesis (in English), Université Paris 6 (2005), available at http://www-drecam.cea.fr/drecam/spec/Pres/Quantro/Qsite/.
- [14]  $\Delta$  varies slightly from contact to contact, possibly due to small differences in the local aluminum film thickness.
- [15] Yu. M. Ivanchenko and L. A. Zil'berman, Sov. Phys. JETP 28, 1272 (1969). V. Ambegaokar and B. I. Halperin, Phys. Rev. Lett. 22, 1364 (1969).
- [16] M. F. Goffman et al., Phys. Rev. Lett. 85, 170-173 (2000).
- [17] D. V. Averin, A. Bardas, and H. T. Imam, Phys. Rev. B 58, 11 165 (1998).
- [18] R. Duprat and A. L. Yeyati, Phys. Rev. B 71, 054510 (2005).
- [19] Both the dc and the ac bias can slightly heat up the on-chip dissipative elements, but in the present case the excess temperature is mainly due to noise (not necessarily thermal) originating in the amplifiers and the biasing sources and which we couldn't completely filter out. This was particularly important for the type A setup.
- [20] S. Shapiro, Phys. Rev. Lett. 11, 80 (1963).
- [21] Contrary to what is observed in large SNS structures, where the physics of fractional Shapiro resonances is governed by non-equilibrium effects. See P. Dubos et al., Phys. Rev. Lett. 87, 206801 (2001); K. W. Lehnert et al., Phys. Rev. Lett. 82, 1265 (1999).
- [22] A. H. Dayem and R. J. Martin, Phys. Rev. Lett. 8, 246 (1962); C. A. Hamilton and Sidney Shapiro, Phys. Rev. B 2, 4494 (1970); O. Hoffmann Soerensen et al., Phys. Rev. B 9, 3746 (1974).

[23] P. K. Tien and J. P. Gordon, Phys. Rev. 129, 647 (1963);
 U. Zimmermann and K. Keck, Z. Phys. B 101, 555